Pis'ma v ZhETF, vol.66, iss.3, pp.195 - 199

LATTICE DEFORMATION FROM INTERACTION WITH ELECTRONS HEATED BY ULTRASHORT LASER PULSE

L.A.Falkovsky, E.G.Mishchenko

L.D.Landau Institute for Theoretical Physics RAS

117334 Moscow, Russia

Submitted 14 July 1997

We propose a model describing destruction of metals under ultrashort intense laser pulses when heated electrons affect the lattice through the direct electron-phonon interaction. The metal consists of hot electrons and cool lattice. The lattice deformation is estimated immediately after the laser pulse up to time of electron temperature relaxation. The hot electrons are described with help of Boltzmann's equation and the equation of thermoconductivity. We use equation of motion for lattice displacements with the electron force included. The estimate of lattice deformation shows that the ablation regime can be achieved.

PACS: 62.20.Fe, 63.20.-e, 79.20.Ds

1. It was pointed out [1], the unltrashort UV laser pulse $\sim 10^{-13}$s results in nonequilibrium electron gas near a metal surface. Such hot electrons were observed in the experiments on IR reflection [2–5], giant electron emission [6–8] and light radiation [9–11] from metals induced by subpicosecond laser irradiation. Because the electron specific heat is much less than the lattice one, these pulses build up the electron temperature T_e considerably higher than the lattice temperature T_i. The characteristic time of electron gas cooling $\tau_e \sim 10^{-12}$s is determined by the electron-phonon coupling constant [12]. The process of temperature relaxation and the subsequent ablation regime have been studied recently [13].

We focus here on the other processes which can take place at times much shorter than τ_e when hot electrons affect directly the lattice expansion through the electron-phonon interaction. This novel effect is caused by the electron gas contribution (depending on T_e) into the elastic constants (sound velocity and optical phonon gap) and owing to the effective electron force proportional to ∇T_e. This interaction induces the lattice deformation.

The experimental results [14] vizualized with the aim of time-resolved X-ray diffraction synchronized with laser pumping can be explained on the framework of the model described below. It was discovered that the laser pumping induces nonstationary increase in lattice parameters of $Au(111)$ and $Pt(111)$ single crystals. The measurement of the shift and intensity variation of Bragg peaks, from the one hand, and observation of the Debye-Waller factor, from the other hand, allows to separate the effects of lattice deformation and heating. It was shown that the initial elastic deformation turns into the subsequent plastic one. Here we propose the theory of lattice deformation due to direct electron-phonon interaction.

2. We describe the overheated electron gas in terms of the Boltzmann equation. When the lattice temperature T_i is higher than the Debye temperature, the mean free of electrons is proportional to $\tau \sim T_i^{-1} \sim 10^{-14}\,\mathrm{s} << \tau_e$. Thus the electron gas is nearly in equilibrium at temperature $T_e >> T_i$. For our problem, the system obeys the equation of thermoconductivity

$$c(T_e)\frac{\partial T_e}{\partial t} + \nabla \mathrm{q} = Q - \alpha(T_e - T_i), \qquad (1)$$

and the equation for the lattice displacement u_i

$$\rho\frac{\partial^2 u_i}{\partial t^2} - \lambda_{iklm}\frac{\partial^2 u_l}{\partial x_k \partial x_m} = G_i(T_e), \tag{2}$$

where $c(T_e)$ is the electron heat capacity, ρ is the metal density, λ_{iklm} is the tensor of elastic constants, Q is the density of the laser energy absorbed by a metal

$$Q(z,t) = I(t)(1-R)\kappa e^{-\kappa z},$$

R being reflection coefficient. Function $I(t)$ describes the pulse shape. The last term in Eq. (1) represents the energy flow from nearly equilibrium hot electrons into the lattice [12]. Constant α is determined by the electron-lattice relaxation $\tau_e \sim \alpha/c(T_e)$. The electron heat flow q and the driving force $G_i(T_e)$ applied to the lattice [15] are defined by the electronic partition function $f_p(\mathrm{r},t)$

$$G_i = \frac{\partial}{\partial x_k}\int \frac{2d^3p}{(2\pi)^3}\lambda_{ik}(\mathrm{p})f_p(\mathrm{r},t), \tag{3}$$

where λ_{ik} is the deformation potential. The effects of the electron interaction (3) on accoustic phonon spectra (as well as on optical phonons) have been studied in detail in our recent paper [16]. The real part of the force (3) (for the first time the corresponding term for optical phonon was written in the paper [16]) leads to the renormalization of lattice parameters - sound velocity for accoustic phonons and optical phonon frequency. The imaginary part results in phonon attenuation due to the electron-phonon interaction. There are two regimes of phonon damping. The first, ballistic one, takes place when electrons are nearly collisionless and the attenuation comes from the electrons moving coherently with the phonon. The hydrodynamic regime is realized when the mean free length of electrons becomes smaller than the phonon wavelength. As a result the attenuation depends strongly on the electron collision rate.

The electronic distribution function in the linearized form

$$f_p(z,t) = f_0(T_e) + \chi_p(z,t)\frac{\partial f_0}{\partial \epsilon} \tag{4}$$

obeys the Boltzmann equation

$$\frac{\partial \chi_p}{\partial t} + v_z\frac{\partial \chi_p}{\partial z} + \frac{\chi_p - \overline{\chi_p}}{\tau} = -\lambda_{ik}(\mathrm{p})\frac{\partial u_{ik}}{\partial t} + \frac{\epsilon_p - \mu}{T_e}\left(\frac{\partial T_e}{\partial t} + v_z\frac{\partial T_e}{\partial z}\right). \tag{5}$$

We write the collision integral in τ-approximation because the electron temperature is much higher than the Debye temperature and electron-phonon collisions have to be elastic. The value averaged over the Fermi surface, $\overline{\chi_p}$ represents the 'in-term' in collision integral. Thorough consideration shows that this term is unimportant when we are interested in thermoconductivity coefficient but it renormalizes the electron-phonon deformation potential: $\lambda_{ik} \to \lambda_{ik} - \overline{\lambda_{ik}}$, (see [17]).

We assume in the following that $T_e << \varepsilon_F$. This let us obtain the results in analytical form. For simplicity we supposed that the metal occupies the half-space $z > 0$, and the boundary conditions to the written above equations have the form

$$\frac{\partial T_e}{\partial z}|_{z=0} = 0, \qquad \frac{\partial u_z}{\partial z}|_{z=0} = 0. \tag{6}$$

The first condition means that the heat flow through the surface vanishes while the second one means the absence of normal stress component.

3. For the times shorter than the electron-lattice relaxation time τ_e, the lattice temperature can be considered as equal to the initial temperature T_0, and the last term in Eq. (1) can be omitted. To solve Eqs. (1) - (5) with boundary conditions (6) we use the even continuation $T_e(z,t)$ and the odd continuation for $u_z(z,t)$ into the half-space $z < 0$.

Let us consider here the solution of Eqs. (1)-(5) for the most interesting case when the times after laser pulse are much longer than the mean free time of electrons ($t >> \tau$) and mean free length is much less than the skin depth ($\kappa l << 1$). Note that electric field of the laser was omitted in Eq. (5) because it is essential only for the times of order of mean free time.

The heat flow in Eq. (1) is calculated with Boltzmann's equation (5). The thermoconductivity coefficient as well as the specific heat becomes proportional to the electron temperatire T_e. The equation (1) turns to be linear in T_e^2 and its solution can be obtaine easily with the help of Green function.

The electron temperature reads

$$T_e^2(z,t) = T_0^2 + \int_0^t dt' \int_{-\infty}^{\infty} dz' \frac{Q(|z'|,t')}{\beta\sqrt{\pi(t-t')D}} \exp\left(-\frac{(z-z')^2}{4(t-t')D}\right), \tag{7}$$

where the diffusion coefficient $D = \tau\overline{v_z^2}$ and the temperature coefficient of electronic specific heat $\beta = c(T_e)/T_e$ are introduced. The function (7) is even in z because $Q(z,t)$ was continued into the $z < 0$ half-space under even manner. Therefore this solution satisfies the boundary condition (6). For the surface $z = 0$ Eq. (7) gives

$$T_e^2(0,t) = T_0^2 + \frac{4}{\pi\beta} \int_0^t dt' Q(0,t-t') e^{\kappa^2 Dt'} \operatorname{erfc}\left(\sqrt{\kappa^2 Dt'}\right). \tag{8}$$

Now let us consider the equation for lattice displacements (2) with the force (3). The main contribution to the force G_i comes from the local equilibrium partition function - the first term in (4), if the condition $t >> \tau$ is valid. Substituting the expression (4) into Eq. (3) and expanding the integral over powers of T_e/ε_F up to the second order, we obtain the force

$$G_i = \Lambda_{ik} \frac{\partial T_e^2}{\partial x_k}, \tag{9}$$

where

$$\Lambda_{ik} = \frac{1}{32\pi} \frac{\partial}{\partial \varepsilon_F} \int \frac{dS}{v} \lambda_{ik}(\mathrm{p}) \sim g\beta,$$

and $g \sim \lambda/\varepsilon_F$ is the dimensionless electron-phonon coupling constant.

To solve the equation of lattice motions (2) with the calculated force (9) from hot electrons, we use the continuation described in the beginning of this section. The function u_z obtains a singularity at $z = 0$ after the continuation. The singularity contributes $d\delta(z)/dz$-term into the second derivative d^2u_z/dz^2. One can use the Fourier transform with respect to spatial coordinate over entire space

$$-\rho(\omega^2 - s^2k^2)u_z(k,\omega) = ik\Lambda_{zz}T_e^2(k,\omega) - i\rho s^2 kC(\omega), \tag{10}$$

where $s = \lambda_{zzzz}/\rho$ is the longitudinal sound velocity in z-direction. The last term $C(\omega)$ has to be determined from the boundary condition (6).

Deformation du_z/dz given by the solution of Eq. (2) can be written as

$$\frac{du_z}{dz} = \frac{i\Lambda_{zz}\kappa(1-R)}{\rho\beta}\int\frac{d\omega dk}{(2\pi)^2}\frac{k^2U(k)I(\omega)}{(\omega+ik^2D)(\omega^2-s^2k^2)}\left(e^{ikz}-e^{i\omega|z|/s}\right)e^{-i\omega t}, \qquad (11)$$

where $U(k) = 2\kappa/(k^2+\kappa^2)$ is the Fourier transform respect to z of laser radiation in metal, $I(\omega)$ is the Fourier transform of the pulse shape $I(t)$. The second term in the brackets corresponds to the general solution of the uniform equation (2) and represents the effect of the surface.

The integrand in (11) contains the poles associated with the diffuson and sound-wave excitations. Sound singularities must be bypassed with insertion of the infinitesimal imaginary term in ω.

4. The electron temperature (7) immediately after the pulse takes its maximum on the surface

$$T^2_{max} \sim \frac{It_0(1-R)}{\beta}\min\left(\kappa, (Dt_0)^{-1/2}\right).$$

This result has a simple explanation. For short pulses $\kappa\sqrt{Dt_0} << 1$ time dependence of temperature corresponds to the local laser intensity in the point of observation. In the opposite case $\kappa\sqrt{Dt_0} >> 1$ the distribution of temperature is determined mainly by the diffusion process.

The lattice deformation (11) vanishes at the surface $z = 0$ according to the boundary condition (6). For $z \neq 0$ the second term in (11) represents the deformation wave propagating from the surface into the bulk of a metal. It gives nonzero contribution only for sufficiently small depths $z < st \sim 10^{-7}$ cm. We see that the maximum deformation takes place at $z \sim 10^{-7}\,\text{cm} << \kappa^{-1}$. It is convenient to perform the integral (11) over ω substituting the Fourier transform $I(\omega)$. We derive the following estimate for the maximum value of lattice deformation

$$\frac{du_z}{dz} \sim \frac{\Lambda_{zz}\kappa(1-R)}{s\rho\beta}\int_0^t dt' I(t')\int_{-\infty}^{\infty}\frac{dk}{2\pi}U(k)\left(\frac{e^{-isk(t-t')}}{s(s+ikD)}-\frac{e^{-k^2D(t-t')}}{k^2D^2+s^2}\right)$$

$$\sim \Lambda_{zz}T^2_{max}\kappa^2t^2/\rho. \qquad (12)$$

We consider times of the order of characteristic time of electron diffusion $t \leq (\kappa^2 D)^{-1} \sim 10^{-12}\,\text{s} << (s\kappa)^{-1}$ but less than the period of sound wave with vavelength $\sim \kappa^{-1}$. Using the estimate $\Lambda/\rho \sim gs^2/\varepsilon_F^2$, we get

$$\frac{du_z}{dz} \sim g\left(\frac{sT_{max}}{\kappa\tau v^2\varepsilon_F}\right)^2 \sim g(1-R)It_0(s/\tau v^2\varepsilon_F)^2/\kappa\beta. \qquad (13)$$

Here we put $s/v \sim 10^{-2}$, $\kappa \sim 10^5\,\text{cm}^{-1}$. and we arrive to the numerical estimate $du_z/dz \sim 10^{-2}g^5(T_e/\varepsilon_F)^2$.

This result agrees with the experiment [14] where a deformation as much as $\sim 10^{-3}$ had been observed. Although our estimate was obtained for $T_e << \varepsilon_F$, it is still correct qualitatively up to $T_e \sim \varepsilon_F$. Therefore, the ultrashort intense laser pulse can result in the destruction and ablation of metals, while the electron component is heated only and the lattice stays cool at considerably low temperature.

In conclusion we would like to emphasize two points. First, as it follows from Eq. (9), the driving force for the lattice expansion is proportional to $T_e \partial T_e / \partial z$. Because of high absorption coefficient of metals in UV region ($\kappa \sim 10^5\,\mathrm{cm}^{-1}$) the temperature gradient reaches $\sim 10^9$ K/cm. Note the extremely high values of this parameter (that is the pecularity of metals) leads to the nonequilibrium expansion of lattice. Second, the subpicosecond elastic deformation of lattice of the order $10^{-3} - 10^{-2}$ corresponding to the internal pressure $10 - 100$ GPa, can provide the effective mechanism for the subsequent laser fracture of metals.

We are grateful to S.I.Anisimov, V.A.Benderskii and B.Rethfeld for many discussions and comments. The work was supported the by Russian Foundation for Basic Research, Grant # 97-02-16044. One of the authors (E.M.) also thanks KFA Forschungszentrum, Juelich, Germany, for Landau Postdoc Scholarship.

---

1. S.I.Anisimov, A.M.Bonch-Bruevich, M.A.El'yashevich et al., Sov. Phys.-Tech. Phys. **11**, 945 (1967).
2. I.G.Fugimoto, J.M.Liu, E.P.Ippen et al., Phys. Rev. Lett. **53**, 1837 (1984).
3. D.M.Riffe, X.Y.Wang, and M.C.Downer, J. Opt. Soc. America B **10**, 1424 (1993).
4. J.P.Girardeau-Montaut and C.Girardeau-Montaut, Phys. Rev. B **51**, 13560 (1995).
5. S.I.Anisimov, V.A.Benderskii, G.Farkas, Sov. Phys. - Uspekhi **20**, 467 (1977).
6. A.G.Krivenko, J.Kruger, W.Hautex et al., Benchte Bunsengeselsch. Phys. Chem. **99**, 1489 (1995).
7. V.A.Benderskii, Russian Electrochem. **33**, 417 (1997).
8. A.G.Krivenko, J.Kruger, W.Hautex et al., Russian Electrochem. **33**, 426 (1997).
9. M.B.Agranat, A.A.Benditskii, G.M.Gandel'man et al., JETP Lett. **30**, 167 (1979).
10. M.B.Agranat, S.I.Anisimov, and B.I.Makshantsev, Appl. Phys. B **47**, 209 (1988).
11. M.B.Agranat, S.I.Anisimov, and B.I.Makshantsev, Appl. Phys. B **55**, 451 (1992).
12. M.I.Kaganov, I.M.Lifshits, and L.V.Tanatarov, Sov. Phys. - JETP **4**, 173 (1957).
13. S.I.Anisimov and B.Rethfeld, Proceedings of "SPIE - The International Society for Optical Engeneering" **3093**, 192 (1997).
14. P.Rentzepis, Abstracts of 2 Conference "Modern trends in chemical kinetics", Novosibirsk, Russia, 1, 142 (1995).
15. V.M.Kontorovich, Sov. Phys. - Uspekhi **142**, 265 (1984).
16. E.G.Mishchenko and L.A.Falkovsky, Sov. Phys. - JETP **107**, 36 (1995).
17. L.A.Falkovsky and E.G. Mishchenko, Phys. Rev. B **51**, 7239 (1995).